Hole spin polarization in GaAs:Mn/AlAs multiple quantum wells

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We study the effect of confinement on the spin polarization of holes bound to Mn acceptors in paramagnetic GaAs:Mn/AlAs multiple quantum wells. It is demonstrated that the polarization of these bound holes is governed by the properties of the host material rather than by quantum confinement. The theory of a single Mn acceptor in a quantum well is developed, and the polarization in the limit of zero temperature is calculated.

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Confinement of carriers in quantum wells (QWs) plays an important role in semiconductor physics because it drastically changes their energy spectrum, renormalizes fundamental quantities such as the effective mass and the g factor, and strongly increases the exchange interaction. One would thus expect that carrier confinement strongly modifies the magnetic properties in the diluted magnetic semiconductor (Ga,Mn)As, which attracts growing interest since its discovery in 1996.[1] However, there have been few studies of either GaAs:Mn/AlAs (Refs. 2 and 3) or (Ga,Mn)As/AlAs multiple quantum wells[4] (MQWs) in the literature, although the study of the spin polarization in these structures allows a direct examination of the origin of ferromagnetism in (Ga,Mn)As. Specifically, the common mean-field model of a free hole gas mediating ferromagnetism via a Ruderman-Kittel-Kasuya-Yoshida (RKKY)-type interaction inevitably results in a complete circular polarization of the hot-electron photoluminescence (HPL) in Faraday geometry and a zero linear polarization in Voigt geometry. This behavior of the HPL polarization is related to the strong anisotropy of the free hole g factor in QWs, which is about zero ($g_{x,y}\sim 0$) in the QW plane and $g\neq 0$ in the growth direction.[5,6] Hence, the equilibrium orientation of the hole spin is accessible in the Faraday geometry, and the HPL polarization can reach a maximum value of unity. In contrast, in the Voigt geometry, no Zeeman splitting of the hole states occurs, and the HPL is unpolarized. Quite different polarization properties of the HPL are expected for models assuming that holes are bound to a Mn acceptor impurity band.[7,8] In this case, the polarization of the HPL is determined by the electronic structure of the ground state of the isolated Mn acceptor,[9] which is sensitive to the confinement as well as to random stress or electric fields.

In this paper, we present a direct study of the hole spin polarization in paramagnetic GaAs:Mn/AlAs MQWs by means of HPL.[7,8] Our study demonstrates that the spin polarization of holes bound to a single Mn acceptor or Mn acceptor impurity band can be increased up to 70% due to confinement in QWs free of random stress and/or electric fields. However, in the experiment the polarization of bound holes is found to be significantly reduced by disorder-induced internal stress and/or electric fields regardless of confinement.

The GaAs:Mn/AlAs MQWs were grown on semi-insulating GaAs(001) substrates by standard solid-source molecular beam epitaxy with an As valved cracker cell. The structures were grown under standard GaAs conditions, i.e., a substrate temperature of 560 °C and a growth rate of about 200 nm/h. All heterostructures consist of 30 periods of GaAs:Mn/AlAs with 10/6 (sample $D1$) and 12.5/3 nm thick wells/barriers (sample $D2$) and a Mn concentration of 5×10^{17} cm^{-3}. A bulklike (1000 nm thick) GaAs:Mn layer with the same Mn concentration (sample $R1$) was grown for reasons of comparison. HPL spectra were measured at a temperature of 5 K and in magnetic fields up to 10 T, either in the backscattering Faraday or in Voigt geometry, using the setup described in detail in Ref. 8.

Figure 1 shows the HPL spectrum obtained from sample $D1$. The sample $D1$ shows near-band-gap photoluminescence centered at 1.441 eV due to the recombination of thermalized free electrons occupying the first subband in the QW with holes bound to neutral Mn acceptors ($1e$-A^0). The lines $2e$-A^0 and $3e$-A^0 are related to recombination of electrons from the bottom of the second and third subbands with holes bound to Mn acceptors, respectively. The band-to-band transition ($1e$-$1hh$) overlaps with line $2e$-A^0. The line $E_g+\Delta$ corresponds to the transition between the first subband of the conduction band and the spin-orbit split valence band.[8] The high-energy cutoff (marked by arrows with label "0") of the HPL spectrum for each sample corresponds to the recombination of electrons from the point of creation with holes bound to single Mn acceptors. A noticeable energy shift (Σ in the inset of Fig. 1) between the excitation energy and the onset 0 of the HPL means that, as in bulk GaAs:Mn,[7,8] the binding energy of the Mn acceptor is close to 110 meV.

Figure 2(a) shows the magnetic-field dependence of the HPL circular polarization (Faraday geometry) measured at its onset 0 for samples $D1$ and $D2$ (note that only spin-polarized holes contribute to the HPL polarization).[7,8] For comparison, we also present data measured on bulk GaAs:Mn (sample $R1$). Evidently, confinement reduces the HPL polarization by a factor of 4 independent of well width. Figure 2(b) shows the corresponding dependence of the HPL linear polarization in Voigt geometry. The linear polarization saturates at $\rho_l < -0.1$ for bulk GaAs:Mn and stays very close to zero (but remains finite) in a QW. These observations contradict the expectation within the RKKY framework.

In the following, we will show that this dependence of the

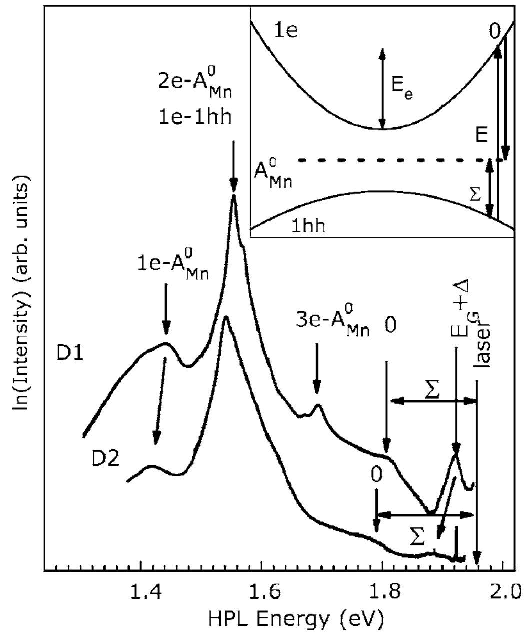

FIG. 1. HPL spectra of samples $D1$ and $D2$ excited with a He-Ne laser (1.96 eV) at $T=5$ K. The arrows labeled "0" indicate the energy for recombination of electrons from the point of generation. The recombination of thermalized electrons in the first, second, and third subbands with holes bound to Mn acceptors is marked by $1e\text{-}A^0_{\mathrm{Mn}}$, $2e\text{-}A^0_{\mathrm{Mn}}$, and $3e\text{-}A^0_{\mathrm{Mn}}$, respectively. The PL band related to recombination of spin-orbit split holes with Mn double donors is labeled $E_G+\Delta$. The inset explains the origin of HPL.

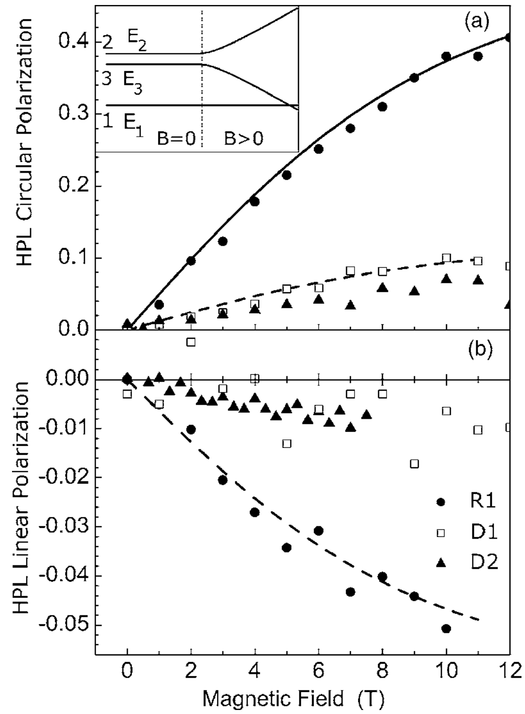

FIG. 2. Magnetic-field dependence of the HPL polarization measured at 5 K in (a) Faraday geometry and (b) Voigt geometry. The solid line is a fit according to Ref. 9 for sample $R1$. The dashed lines are guides for the eyes. The inset in (a) represents the sublevels of the $F=1$ ground state split due to random fields and confinement in accordance with Eq. (4).

HPL linear and/or circular polarization in the Voigt and/or Faraday geometry on the confinement in MQWs can be understood within a model for holes bound to single Mn acceptors in GaAs:Mn.

To demonstrate this, we study the effect of confinement and random fields on the HPL polarization (in the low-temperature limit $T=0$) of a single Mn acceptor located in the center of a QW. In GaAs, Mn predominantly incorporates substitutionally on a Ga site and thus gives rise to an acceptor level at about 0.11 eV above the valence band. The electronic state of the Mn acceptor is determined by the Mn^- core, which in spherical approximation behaves as an $S=5/2$ state. The hole in the $1S_{3/2}(\Gamma_8)$ state is characterized by an effective angular momentum $J=3/2$.[9] The total angular momentum of the system F varies from $|S_d-J_h|=1$ to $S_d+J_h=4$. Due to the exchange interaction between holes and $3d^5$ electrons, the single level splits into four sublevels: $F=1,2,3,4$, with $F=1$ being the ground state for antiferromagnetic exchange.[9,10] The interaction with the magnetic field leads to the splitting of the $F=1$ state into three sublevels with an energy

$$E_{F=1}=\mu_B g_{F=1} m_{F=1} B, \tag{1}$$

where $m_{F=1}=0,\pm 1$ is the angular momentum projection and $g_{F=1}$ is the g factor of the ground state (the wave functions and the g factor $[g_{F=1}=(7g_e+3g_h)/4=11/4$, where $g_h=-1$ and $g_e=2$ are the heavy hole and $3d^5$ electron g factors, respectively] of the ground state are given in Ref. 9).

The exchange interaction strongly modifies the selection rules for the $e\text{-}A^0_{\mathrm{Mn}}$ optical transition. Particularly, the circular polarization of emitted light for a strong magnetic field ($\mu_B g_{F=1} B \gg kT$) is reduced by the exchange interaction in bulk GaAs from 1 to 5/7.[9] Random local electric or stress fields (δ) acting on the Mn acceptor states result in a further reduction of the HPL circular polarization due to a splitting of the $F=1$ ground state.[9,10] The typical value of this splitting for Czochralski-grown GaAs:Mn was directly measured by means of Raman scattering to be $\sim$1 meV.[11] For this splitting, the HPL circular polarization saturates at $\approx$0.5 (see Ref. 9) in agreement with experimental data.[7,10] With increasing strength δ of these random fields, the HPL circular polarization varies from $\approx$0.5 to $\approx$0.26, as illustrated by the solid line in Fig. 3(a) obtained by the model developed in Ref. 9 with the assumption that only the lowest energy state is occupied by holes. The dependence of the linear HPL polarization, calculated for the Voigt geometry in the same model, is shown by the solid line in the inset of Fig. 3(b).

In a QW, confinement is expected to compete with these random fields and thus to enhance the polarization of the $e\text{-}A^0_{\mathrm{Mn}}$ transition. Indeed, the ground state of a shallow acceptor splits into light- and heavy-hole subbands in a QW, with the latter being the ground state. The light hole–heavy hole splitting depends on the well width and varies for a shallow acceptor (Be) in the range of $\Delta\sim 2$–7 meV for

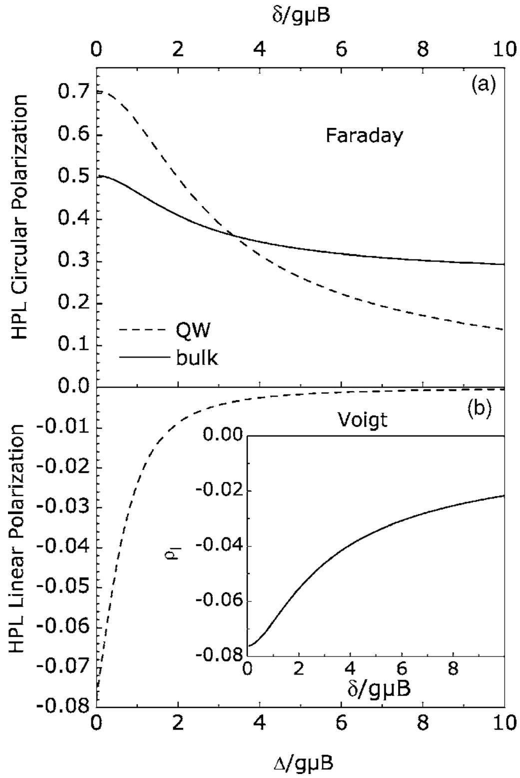

FIG. 3. The calculated dependence of the HPL circular (a) and linear (b) polarizations on random fields (δ) and light hole–heavy hole splitting (Δ) induced by confinement. The inset presents the calculated dependence of the HPL linear polarization on the strength of random fields δ in the bulk material.

widths of 10–4 nm.[6] The energy splitting caused by confinement may be calculated using the zero potential method.[12] This approach yields a value of $\Delta \sim 0.5$–5 meV for a well width of 10–5 nm in GaAs QWs for acceptors with a binding energy between 50 and 100 meV. The Hamiltonian describing the confinement effect on acceptor state is

$$H_{\Delta} = \Delta(\hat{F}_z^2 - 2/3), \tag{2}$$

where F_z is the z projection of the total angular momentum (z coincides with the growth direction). The influence of random fields may be described by the Hamiltonian

$$H_{\delta} = \delta(\hat{F}_X^2 - 2/3), \tag{3}$$

where δ is the splitting caused by the random fields which are assumed to be directed along the XY plane in a QW. We conjecture that in QWs grown at high temperature, the random fields originate from interstitial Mn acting as ionized donors rather than from random stress. Even for standard growth conditions, it has been demonstrated recently that a fraction of the Mn is indeed incorporated as ionized interstitials.[13] The simultaneous influence of size quantization and random fields is described by the sum of Eqs. (2) and (3). For $B=0$, the energies of the sublevels are given by

$$E_1 = \frac{-2\Delta + \delta}{3}, \quad E_2 = \frac{\Delta + \delta}{3}, \quad E_3 = \frac{\Delta - 2\delta}{3}. \tag{4}$$

The finite polarization observed for the bulk samples, together with the fact that the heavy-hole subband is the ground state of QW, implies that both δ and Δ are positive. Thus, a level scheme results as depicted in the inset Fig. 2(a). The two types of perturbations (2) and (3) acting on the single Mn acceptor distinguish the present case of QWs from the bulk. To demonstrate this, we calculate the HPL polarization in the low-temperature limit[14] using the approach developed in Ref. 9.

In the Faraday geometry, and when only sublevel "2" [see inset of Fig. 2(a)] with energy E_2 is occupied by holes (i.e., in the low-temperature limit $kT \ll g\mu_B B, \delta, \Delta$), the degree of the circular polarization can be expressed as

$$\rho_c = \beta \frac{5\epsilon^2 + 2\epsilon(5\gamma_1 - 4\gamma_2) + 5\gamma_1^2 - 8\gamma_1\gamma_2 + 5\gamma_2^2 - 9\gamma_3^2}{7\epsilon^2 + 2\epsilon(7\gamma_1 - 4\gamma_2) + 7\gamma_1^2 - 8\gamma_1\gamma_2 + 7\gamma_2^2 + 19\gamma_3^2}, \tag{5}$$

where $\epsilon = Em_0/E_e m_e$, E_e is the kinetic energy of electrons in the conduction band, E is the PL photon energy (see inset in Fig. 1), m_e is the electron effective mass, and γ_1, γ_2, γ_3 are the Luttinger parameters. Expression (5) was obtained for the recombination of hot electrons with the wave vector lying in the QW plane, i.e., $k_x = k_y = k/\sqrt{2}$ and mean value of $k_z \sim 0$. As expected, ρ_c does not depend on Δ because in the Faraday geometry the projection of the acceptor angular momentum coincides with the magnetic field applied parallel to the QW growth direction.

The effect of random fields in Eq. (5) is taken into account by the parameter $\beta = 1/\sqrt{(1 + (\delta/2\mu_B g_{F=1} B)^2}$. Evidently, the circular polarization decreases with an increase of δ, i.e., an increase of the strength of random electric and/or stress fields in the XY plane of the QW. For example, using the parameters of our experiments ($E \sim 1.8$ eV and $E_e \sim 0.3$–0.4 eV), Eq. (5) predicts ρ_c to decrease from 0.5 to 0.3 when $\delta/2\mu_B g_{F=1} B$ increases from 1 to 2. The dependence of the HPL circular polarization on in-plane random fields δ is shown for the present experimental case by the dashed line in Fig. 3(a). When comparing this result to the experimental one [cf. Fig. 2(a)], it becomes clear that random electric fields determine the hole spin polarization in the QW (as well as in the bulk[8]) to a very large extent. For the field-free case ($\delta = 0$), the circular polarization is significantly higher for QWs compared to the bulk. However, for $\delta \to \infty$, the circular polarization is reduced to 0.26 for the bulk, while the polarization approaches zero for QWs. As mentioned in Ref. 14, the actual polarization will be even lower than these values when finite temperatures are considered.

In the Voigt geometry [see Fig. 2(b)], confinement *decreases* the degree of the linear polarization in QWs free of fields. For the lowest sublevel occupied by holes, the degree of the linear polarization can be written as

$$\rho_l = \frac{2\alpha^2[3\gamma_3^2 - (\epsilon + \gamma_1 + \gamma_2)(\epsilon + \gamma_1 - \gamma_2)]}{2(19\alpha^2 + 11)\gamma_3^2 + (11\alpha^2 + 4)(\epsilon + \gamma_1 - \gamma_2)^2 + (3\alpha^2 + 2)(\epsilon + \gamma_1 + \gamma_2)^2}, \tag{6}$$

where

$$\alpha = \frac{1}{\sqrt{2}}\left[\frac{\Delta}{\mu_B g_{F=1} B} - \sqrt{1 + \left(\frac{\Delta}{\mu_B g_{F=1} B}\right)^2}\right]. \tag{7}$$

In the bulk, $\alpha = -1/\sqrt{2}$ and $\rho_l \approx -0.078$, in good agreement with the experiment [see Fig. 2(b)]. In QWs, the effect of confinement reveals itself in the significant decrease of ρ_l (cf. Fig. 2). The origin of this decrease in polarization for QWs compared to the bulk case is the confinement effect contained in Eq. (2). The dependence of the HPL linear polarization on confinement (Δ) for the experimental value of the hot-electron kinetic energy and $\delta = 0$ is presented by the dashed line in Fig. 3(b). Equation (6) predicts $\rho \sim -0.024$ for $\Delta/\mu_B g_{F=1} B = 1$. Finite random fields ($\delta > 0$) applied in the XY plane can only decrease the linear polarization similar to the random stress in the bulk case [see inset in Fig. 3(b)]. Again, and as in the case of the Faraday geometry, the actual polarization at finite temperatures will be even lower than these values when the occupation of all levels are considered.[14]

In conclusion, our model of holes bound to single Mn acceptors in a QW shows that confinement does *not* increase the spin polarization of holes *per se*, contrary to expectation. The magnetic behavior even of narrow QWs is governed by random stress and/or electric fields in the QW as in the bulk, and is not enhanced by confinement. The latter conclusion is important for an understanding of the polarization properties of dilute (Ga,Mn)As QWs, the random fields are dominating and drastically reduce the polarization even for the bulk. Using our model, one can explain the fact that in dilute (Ga,Mn)As QWs the easy axis of magnetization lies in the QW plane[4,15] but not in the growth direction as expected in the RKKY-type model.

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[14] A general description at $T \neq 0$, is complicated by the fact that one has to also consider transitions to sublevels "1" and "3," whose polarization depends on the strength of δ, as well as on $\mathbf{B}$. For $z \parallel \mathbf{B}$, the radiative recombination of electrons with holes located on sublevel 1 gives rise to $\rho_c^1 = 0$, while the transition to sublevel 3 is accompanied by a sign reversal of the polarization ($\rho_c^3 = -\rho_c^2$). Particularly, in Faraday geometry, when $\delta \perp \mathbf{B}$ and $\delta \gg \Delta, g\mu_B B$, $\rho_c^3 = -\rho_c^2 \to 0$. Taking into account these facts, the actual polarization will thus always be lower than the one calculated here.

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